A REVIEW OF WASTE TYRE PYROLYSIS: A DISCUSSION AND ANALYSIS

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ABSTRACT

Over the years, the prospective of end-of-life tyres as a source of fuel, a chemical feedstock and carbonaceous material through pyrolysis has been explored immensely. Through literature review evaluation and analysis, this paper aims to present and discuss the development of waste tyre pyrolysis process as well as the derived products with their possible application pathways. The various reactors that can be employed during the pyrolysis process are discussed. Furthermore, the different types of pyrolysis processes as well as their influence on product yield are highlighted. The paper drew the conclusion that the pyrolysis literature which has been continuously developed over the years emphasize the need to fully understand how operating conditions such as temperature, pressure, heating rate, retention time, feed size; material feed rate ; reactor selection and the type of pyrolysis influence the selection and optimization of product stream/s.

Key Words: *Reactor Selection, Types of Pyrolysis Products, Tyre derived Products, Waste Tyre Pyrolysis.*

NOMENCLATURE

Symbols :	Heatinng rate, $^oC/min$
Temperature, ^oC	Mass flow rate, kg/hr
Pressure, kPa	Weight percenrage,wt%
Retention time, hr	Particle size, cm

1. INTRODUCTION

The increasing standards of living and value-added industrialization are showing an increase in the amount of automobile users. As a consequence, the car generation statistics are increasing globally, for example, the European Tyre and Rubber Manufacturers Association (2015) has reported that the projected quantities in Europe for 2025 can possibly arrive at 347 million cars when compared with the 322 million cars manufactured in 2014 [1]. Similar cumulative trends are realized for China and India where more than a two-fold increase is expected in 2025, 332 and 69 million cars respectively. An annually increasing trend is predicted for most first-world counties such as the United States (US) and the European Union (EU), furthermore, developing counties such as South Africa [2]; Brazil; Taiwan and Indonesia [3] are significantly adding to the global waste generation figures. Additionally, the numbers of trucks seen on the roads are also growing rapidly due to growing economies leading to a higher demand for products. Truck tyres have relatively different properties compared to passenger car tyres for example durability which makes their disposal difficult.

2.THE WASTE TYRE PROBLEM

Tyre vulcanization serves to enhance its elasticity and durability. Antioxidants and anti-ozone reagents such as sulphur are supplemented to tyres to prolong their working lifespan. The vulcanization process makes tyres non-

degradable [4]. Rubber is abrasive and as well as water, electrical, heat, chemical and biological resistant. Microorganisms would require more than 100 years to destroy tyres [5]. Due to their non-degradability waste tyres gather in bulks to be incinerated or stockpiled in landfill sites. When tyres are incinerated in public and unprotected spaces, they release toxic carcinogens [6,7]. Global pollution regulations have compelled tyre incineration to be operated under controlled environments [2]. However, this is associated with pricey air monitoring and regulating system.

The prime factor that contributes to scrap tyres being bulky and inhabiting large quantities of valuable space is their void space, which constitutes approximately 75%. Furthermore, stockpiled tyres and landfills gases such as methane form a volatile mixture that can cause ground and water pollution. Discarded tyres also pose as propagation sites for vermin [8,9]. Mosquitoes are associated with deadly diseases and viruses like zika. Lastly, waste tyres also cause serious environmental threats due to their fire hazard resulting in air pollution [10]. These fires tend to be widespread and uncontrollable because of the high flammability property as well as the high calorific value associated with tyres.

3. PYROLYSIS

The global dependence on fossil fuel is decreasing due to diminishing supplies. On the contrary, the demand for energy is projected to increase five times more by 2021 [11]. Consequently, the development of alternative waste to energy initiatives have gained world traction. Pyrolysis is perceived to be a novel technology that generates valuable end-products from end-of-life tyres. Pyrolysis is an energy induced process that promotes the thermal breakdown of components in the absence of spontaneous gases, such as air or oxygen. Four conventional products are produced during the pyrolysis of waste tyres, namely: gas, liquid (oil), solid (char) and steel wires. A detailed schematic diagram of the waste tyre primary and secondary valuable products is presented in Figure 1. The process efficiency is approximately 70%, however, efficiencies of 90% can be achieved with the use of gas product to sustain the process energetically [12]. Likewise, tyre shreds can be utilized to enhance process efficiencies by 20-30% [13]. Research has shown that pyrolysis fundamentally takes place at temperatures between 400 and 800°C [14, 15, 16, 17]. They have also shown that product distribution is strongly influenced by the variation in temperature. J. D. Martinez et.al (2013) highlighted that pyrolysis processes operated under lower temperature conditions usually favours the production of excess liquid products whereas higher temperatures favour the generation of gas [16]. Feed size, reactor type, heating rate, vapour residence time and pressure are important parameters that affect the system especially the final product yield, this is further emphasized by R. M. Islam (2010) [12].

Pyrolysis Process	Retention time (hr)	Heating rate (°C/min)	Particle size (cm)	Temperature (°C)	Product yield (wt%)		
					Oil	Char	Gas
Slow	0.125–0.153	0.02-0.22	0.5-5	275–675	30	35	35
Fast	0.00139–0.0028	2.2-44	<0.1	575–975	50	20	30
Flash	<0.00139	>219.7	<0.02	775–1025	75	12	13

TABLE 1. Types of pyrolysis processes

3.1 Types of pyrolysis processes

Different kinds of pyrolysis processes have been documented in literature by M. I. Jahirul et.al (2012), namely; atmospheric, vacuum, catalytic, fast, ultra-fast (flash) and slow [18]. Table 1 shows the classification of the pyrolysis processes with their operating parameters and resulting product distribution quantities. From Table 1, it is evident that fast and flash pyrolysis at high operating temperatures favours the production of oil products, due to short residence times that allows for fast volatilization followed by rapid condensation.

3.3.1 Slow pyrolysis

M. I. Jahirul et.al (2012) showed that slow pyrolysis of waste tyres at low temperature and heating rate is predominantly employed to advance the production of char, [18], further shown in Table 1. This process is characterized by prolonged residence times in which the gas phase products further react ensuring the generation of excess solids. However, D. Ratnakiran (2017) highlighted the shortcomings of slow pyrolysis, namely; the

substandard quality of oil produced during the process, the reduced oil yield [19] and excess energy requirement due to prolonged residence times [19, 20].

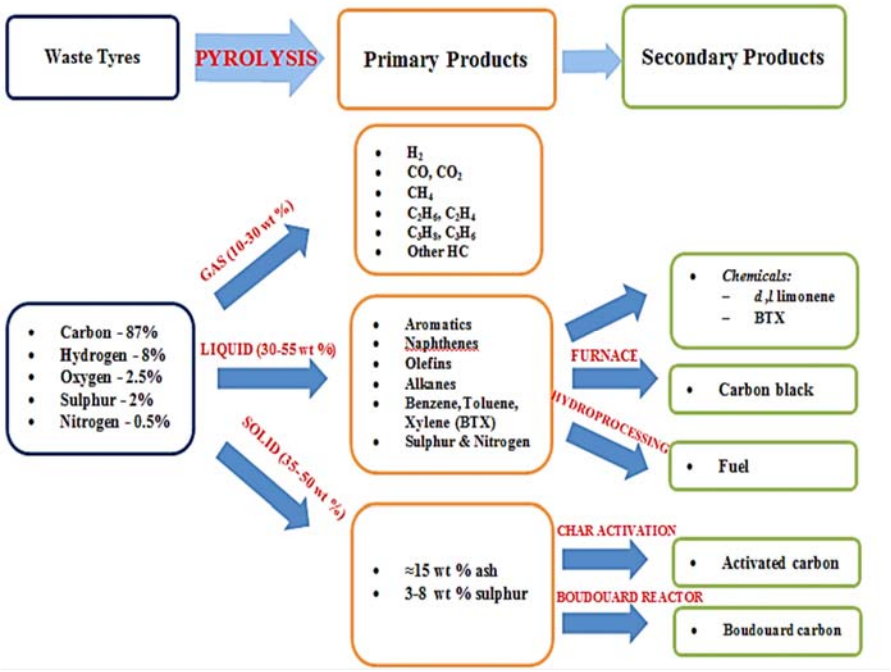

FIGURE 1. Waste tyre pyrolysis derived products

3.3.2 Fast pyrolysis

This process is characterized by the prompt volatilization of material at high temperatures. Fast pyrolysis has the potential of producing 60%–75% liquids, 15%– 25% solids and 10%–20% gas products [18]. The fundamental properties of fast pyrolysis are comprehensively discussed by N. Tippayawong et al, (2008) [21], where elevated heat transfer and heating rates, quick vapour retention time, prompt gas cool off times and accurate reaction temperature controls are the major characteristics highlighted. Fast pyrolysis is highly recommended for liquid fuel production and a variety of valued industrial chemicals. The process has been perceived by Demibas (2002) to require low capital investment and produce high energy competences when likened with other parallel processes [22].

3.3.3 Flash pyrolysis

This process is associated with a high heating rate and thermal conditions as well as extremely quick gas retention times. However, this process has presented some shortcomings, for instance: temperature unsteadiness and the production of a corrosive prone liquid, the existence of solids in the liquid and the generation of water in the process [23]. Atmospheric pyrolysis is achieved under ambient pressure conditions while vacuum pyrolysis is carried out under an extremely low-pressure environment, generally at 4,0 kPa [24]. Roy and Chaala (1999) used shredded waste tyres in a vacuum pyrolyzer and proved that vacuum pyrolysis showed to be a promising process for energy reclaiming [2].

3.3.4 Catalytic pyrolysis

Pyrolytic catalysis has been reported to enhance product yields or advance the product properties to noteworthy industrial applications. Numerous researchers have investigated the impact of catalysis on tyre pyrolysis and the end products. Olazar et al. (2008) studied the influence of catalysis at an operating temperature range of 425-500oC. The results showed that pyrolyzing waste tyres in the presence of Zeolite Socony Mobil–5 (HZSM5) catalyst improved carbon black production from 34.0 to 45.0%, gas production from 8.0 to 20.0% and decreased tar production substantially from 28.8 to 14.9% [25]. Moreover, in HY Zeolite catalyst, char yield was marginally increased,

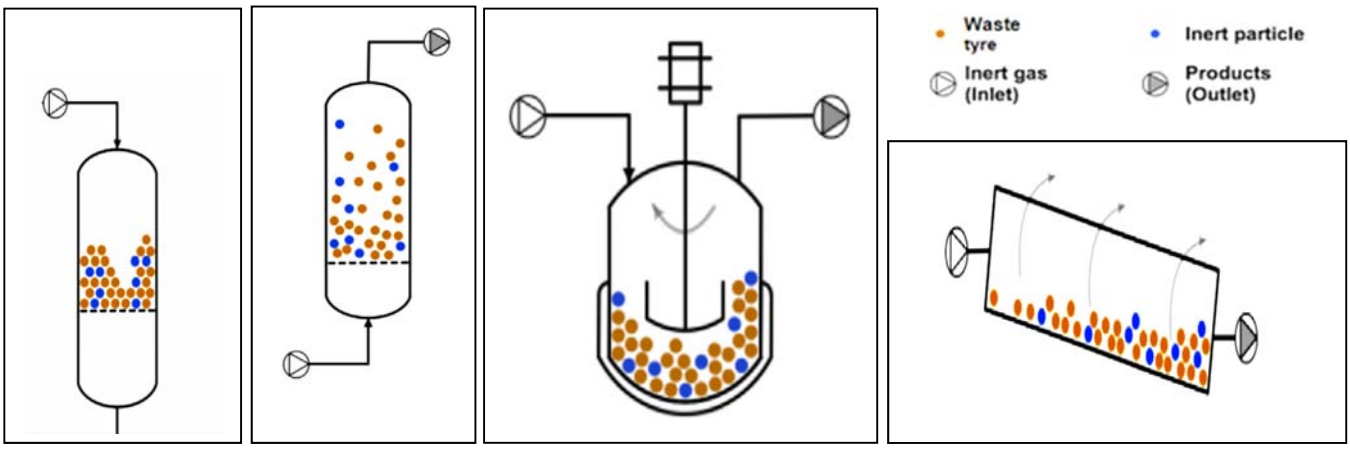

FIGURE 2. (a) Schematic of a fixed bed reactor, (b) Schematic of a screw kiln and rotary kiln reactors, (c) Schematic of a fluidized bed reactor (d) Schematic of a stirred tank reactor

while a reduction in liquid yield was noted, no changes were observed in the gas yield, and tar yield increased significantly from 42.7 to 32.5% [25].

Shah et al. (2008) investigated the effect of catalyst at 350^0C and atmospheric pressure conditions. He reported that MgO catalyst favoured the production of pyrolytic oil with an optimum yield of 39.8% [26], while CaCO3 promoted the generation of vapour product with an optimum yield of 32.5% [26].

Kar (2011) studied the impact of catalyst quantity on pyrolysis product yield. The results showed that supplementing the process with 5% perlite at 3750^0C upgraded pyrolytic oil production from 46.2 to 62.0%, gas production from 3.0 to 8.0% and reduced solid production from 50.7 to 28.0%. In addition, raising the catalyst amount from 5 to 10% improved oil production from 62.0 to 65.0% [27].

3.2 Types of pyrolysis reactors

There has been a large range of reactors developed over the years for the application of waste tyre pyrolysis and the most generally used are fixed-bed (batch), screw kiln, rotary kiln, vacuum and fluidised-bed [28]. Reactor design has been found to be one of the paramount factors that affect product generation, gas and oil characteristics, temperature and heating rate selection. William (2013) summarized the different types of pyrolysis reactors developed over the years by various researchers and also presented their different product yields and experimental conditions [29].

3.3.1 Fixed bed reactor

These reactors are commonly utilized for slow pyrolysis in batch systems [28], a representation of a fixed bed reactor is shown in Figure 2(a). The wall of the reactor chamber is normally heated externally, nitrogen or an alternative inert gas is purged through the system and act as carrier gas. Waste tyre pyrolysis tests are normally carried in the temperature range of 450-700 °C due to fact that their thermal degradation typically resumes at approximately 350 ºC [28]. These reactors may comprise of a catalyst, normally in tablet form in a stationary position. This serves as a major restraint as this hinders contact with the catalyst [29]. The generated gas is cooled and condensed as a liquid, the non-condensable gases support the process and the solids are collected upon completion of the process.

The effect of tyre category and composition on product yield in a fixed bed reactor was studied by Ucar et al. (2005). At temperatures of 550, 650 and 850 ºC under a nitrogen carrier gas, the obtained gas composition from both car and truck tyres were comparable. The physical properties of the oils were similar, however, truck tyres showed lesser sulphur and aromatic content when compared to passenger tyres [30]. In literature, Rowhani and Thomas (2016) reported an inverse relationship between the natural rubber to synthetic rubber composition in passenger and truck tyres (14% and 27% of natural rubber and synthetic rubber respectively make up the total rubber used in passenger tyres while the inverse is noted for truck tyres). Synthetic rubber, which makes up 60%

of the rubber used in a tyre formation is a petrochemical derivative in a form of styrene-butadiene and polybutadiene [30]. Therefore, this complex mixture could be responsible for the higher aromatic yield in passenger tyres as compared to truck tyres. Moreover, the authors also reported lower sulphur content in the ultimate analysis of truck tyres (1.4wt%) in relation to passenger tyres (1.7wt%).

Islam et al. (2011) studied the combined pyrolysis of bicycle, passenger and truck tyres at in a fixed-bed reactor under the temperature range of 375 to 575ºC and obtained variable oil yields from 45 wt % to 56.5 wt % [31]. At temperatures higher than 575ºC, the oil yield reduced to 8 wt % and the gas yield obtained was 20 wt %. This is attributed to the fact to the rapid volatilization resulting in increases in gas yield and decreases in oil yield at higher temperatures.

Industrially applied fixed bed batch reactors for pyrolyzing tyres are well established, however, the need for a continuous system has led to the study of rotary kiln, moving bed screw, fluidised bed, stirred tank and conical spouted bed reactors where tyres are fed immediately to the reactor.

3.3.2 Screw kiln and rotary kiln reactors

A rotary kiln reactor is a cylindrical, refractory material lined reactor and is slightly inclined (1–10°) to progress the waste material forward, shown in Figure 2(b). The char is retrieved at the reactor ending. As an added advantage, the process speed of turning, extent of filling and particle dimensions can be optimized to improve product yield. Furthermore, rotary kilns permit a wide variety of particle dimensions and a range of material for co-processing [32]. Li (2004) [33], Antoniou and Zabaniotou (2015) [34] and Galvagno (2002) [35] conducted studies into the impact of temperature in a rotary kiln reactor. Waste tyre pyrolysis experiments were carried out at a temperature of 600 °C and obtained 18 % [33], 10.80 % [34] and 8.16 % [35] of gas yields respectively. The data obtained by the researchers show varying results under the same conditions.

A comparison study of a fixed-bed and rotary kiln reactors into the influence of reactor type, product yields were conducted by Aylón et al (2008) [36]. The results show that both reactors performed optimally at 600 ºC under an inert environment. A throughput of 15kg/hr was employed in the rotary kiln reactor. The results show 100% waste tyre conversion for both reactors, however, the fixed-bed reactor demonstrated a longer retention time. Char yields of up to 38 wt % were obtained for both reactors, the oil yield was reduced from 54.6 to 43.2 wt % and the gas yield was raised from 7.5 to 17.1 wt % in a rotary kiln reactor.

3.3.3 Fluidized bed reactors

Fluidized beds are commonly employed in the fast pyrolysis process and require small particle sizes [16] and quarts sand is typically used as the fluidizing bed medium [2], a schematic representation of a fluidized bed reactor is presented in Figure 2(c). The fluidized bed is heated indirectly by the pyrolysis gas, furthermore, nitrogen and steam can be utilized as a fluidizing gas.

Kaminsky et al. (2009) researched the effect of temperature on truck tyres in a fluidized bed reactor [37]. The outcomes show that raising the temperature from 700 to 880 ºC increased gas yield, decreased oil yield, and the char production remained uniform. The product yield was not altered when the material feed rate was raised from 0.321 to 0.52 kg/hr.

A laboratory-sized fluidized-bed reactor was utilized by Kaminsky and Mennerich (2001) to investigate the effect of temperature and fluidizing agent on product generation where synthetic tyre rubber was used as feed material [38]. Quartz sand was the fluidizing bed medium of choice. Temperatures of 500, 550 and 600 ºC and a gas retention time of 2.6-3.0s were employed. When nitrogen was used as the fluidizing gas, char yield of 30.00-40.00 wt%, oil yield of 51.24-65.59 wt% and gas yield of 5.0-9.2 wt% were produced correspondingly. For the second study, steam was employed as a fluidizing gas at 500 ºC, as a result, a char yield of 31.0 wt%, tyre-oil yield of 65.0 wt% and gas yield of 3.5 wt% were obtained. It can be deduced that comparable product yields are obtained from the use of nitrogen and steam as fluidizing agents.

3.3.4 Stirred tank reactors

These reactors are built on an autoclave type design, where waste tyres and the inert gas are fed continuously. Stirred tank reactors are designed for processing whole tyres, resulting in a considerable saving on size reduction costs [39]. Figure 2(d) represents the schematic design of a stirred tank reactor. Rodriguez et al. (2001) conducted tyre pyrolysis in a stirred tank reactor under the following conditions: reactor volume of 3.5dm^3, nitrogen inert

conditions, tyre particle sizes of 2-3 cm, sample quantity of 175g, temperatures of 300, 400, 500, 600 and 700 ºC, heating rate of 15 ºC/min and a retention period of 30 min [2]. The resultant products char yield of 43.7-84.7 wt%, tyre-oil yield of 4.8-38.5 wt% and gas yield of 7.7-19.3 wt%. Rodriguez et al. concluded that pyrolysis temperatures of 500, 600 and 700 ºC showed complete decomposition of tyres. Also, 500 ºC proved to be the optimum temperature for tyre oil production.

3.3.5 Conical spouted bed reactors

The CSBR reactor was initially studied in batch operation by Olazar et al. (2005) [40], Aguado R. (2000) and Aguado et.al (2003) studied the continuous operation for other wastes while López et.al. (2009) [43] applied it for waste tyres. An example of the CSBR reactor is shown in Figure 6. The co-pyrolysis of natural and synthetic rubber was carried out at various temperatures namely; 425, 500 and 600 ºC. Natural rubber yielded the following results; char 36-38.30 wt%, tyre-oil 54.88-62.83 wt% and gas yields of 1.81-6.81 wt% while the product compositions for synthetic rubber were 33.91-35.81 wt%, 33.91-35.81 wt% and 1.81-8.26 wt% respectively.

Furthermore, [43] concluded that the carbon black generated at 600 ºC was of activated carbon grade. The results obtained by Lopez et al (2009) are attributed to the attractive features of CSBRs which are a modest design (gas distributor not required); low-pressure drop; accommodates a variety of feed material and sizes and short retention times of the gases in the reaction chamber [44].

The literature studies presented and analysed indicate that the design of a reactor influence critical aspects such as the disintegration of waste tyres, product yield and product composition. Additionally, when selecting a

pyrolysis reactor economics, kind of feedstock, desired product mix, resources and other operating factors should be considered.

3.3 Types of pyrolysis reactors

Waste tyre pyrolysis generates four product streams namely: gas, liquid (oil), solid (char) and steel wire. The three mainstreams oil, char and gas are discussed.

3.3.1 Gas

The gaseous portion of waste tyres pyrolysis products consists mainly of C_1–C_4 hydrocarbons, H_2, CO, CO_2, with minute amounts of H_2S, SO_2 and NH_3 [45,46]. Pyrolysis gas can be utilized to self-sustain the pyrolysis process, thus, proving to be energy-efficient [47]. H_2S is formed from the release of sulphur compounds into gaseous form during pyrolysis. This is the sulphur used in the vulcanization step during the fabrication of tyres. Its concentration in pyrolytic gas is usually minimal. The heating value of pyrolytic gas has been reported in literature to be 42 MJ/kg, this is a considerably high value when linked with natural gas (35-40 MJ/m^3 depending on the source of origination) [48]. As a result, pyrolytic gas seems to show great potential when regarded as a source of fuel [48]. The energy reclaimed from the gas can be utilized for other noteworthy applications besides pyrolysis. However, the SO_2 concentration in the exhaust gases are greater than the regulatory limits [15]. Consequently, gas cleaning control measures for the removal of harmful substances will be required in order to meet regulatory requirements.

3.3.2 Liquid

The liquid fraction is a composite blend of alkane and alkene series as well as aromatic compounds [11] and constitute to a very high heat value between 40–45 MJ kg−1 [29], akin to that of petrol and diesel (44−46 and 42−46 MJ kg−1 correspondingly) [49]. The high sulphur concentration brings about challenges in utilizing the oils as fuel. Diesel has a comparable calorific value to that of pyrolytic oil; however, the sulphur concentration is significantly low, about 0,1 wt% [49]. Numerous techniques for sulphur removal are well documented in literature, e.g. dosing chemicals such as CaO, $Ca(OH)_2$ or NaOH, further refining the oil, oxidizing the oil with H_2O_2 in combination with an acidic catalyst or hydro-refining [15].

Another possible pathway that can be employed to make waste tyre pyrolysis more attractive is the recovery of valuable chemicals. The liquids consist of a variety of beneficial chemicals, such as dipentene; BTX; ethylbenzene etc. [49]. Dipentene consists of two enantiomorph compounds namely; d- and l-limonene. Rodriguez (2001) documented the properties of dipentene comprehensively at ambient conditions. Furthermore, in 2017 Czajczyńska et al. (2017) evaluated the value of dipentene to be US$1500-US$2500 per metric tonne, depending on its purity [15]. It is primarily employed as an industrial solvent, cleaning agent for, e.g., electrical

circuit boards, as a dispersing agent for pigments, or as a constituent in pesticide products [16]. d-Limonene has been reported to have an orange-like aroma, while l-limonene has a pine aroma. Their distinctive smells allow for them to be employed for fragrances and flavourants, on condition that the two enantiomorphs can be effectively splitted.

3.3.3 Carbon black

The carbon black/char can be utilized instantly as a fuel in a form of briquettes, or post enhancement as an activated carbon or carbon black [50, 51]. This fraction exhibits a permeable material with a typical heating value of 30 MJ/kg. The composition and characteristics of the char are dependent on the pyrolysis parameters and the waste tyre composition. Pyrolytic char can be further enhanced chemically and physically producing valuable secondary products. To yield a chemically activated carbon, char is saturated with concentrated aqueous solutions of an activation agent, of which $ZnCl_2$, KOH and H_3PO_4 are frequently used. Chan et al. (2011) came to the conclusion that there is a direct proportional relationship between the activation agent and the quantity of dosage, however, the level of activation is also defined by the choice of chemical, the intensity of the mixing, temperature and the length of the activation step [52]. Physical activation is the partial oxidation of carbon-based material to intensify its adsorption capacities and is commonly carried out as a secondary phase after carbonization [53]. A two-phased process has been developed. During the initial phase of activation unstructured carbon-like tar is incinerated with the aim of opening blocked pores, moreover, carbon crystallites are oxidized during the second phase with the aim of increasing the surface area and the creation of new micropore space [53]. Conventional gases used during physical activation are steam/H_2O, CO_2 and air/O_2 [54].

Some of the predominant markets for pyrolytic char are briquettes, fuel for boilers or as activated carbon. Pyrolysis derived activated carbon has the potential to be utilized for industrial and municipal wastewater purification techniques [55].

4. CONCLUSIONS

The pyrolysis of waste tyres has offered a worthwhile solution for the reprocessing of end-of-life tyres. However important factors such as reactor selection, pyrolysis process type (commonly, slow, fast and ultra-fast pyrolysis), process conditions (temperature, pressure, heating rate, residence time, feed size, material throughput etc.) and the desired product stream should be considered when evaluating an investment in waste tyre pyrolysis. In addition, the economics and viability of the chosen stream should be explored and well researched.

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The authors are grateful to the University of Johannesburg's Global Excellence and Stature scholarship and conference funding, the Chemical Engineering Department at the University of Johannesburg for the continued support and encouragement. The Botswana International University of Science and Technology is also acknowledged for supporting this research.

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